This Page Is Inserted by IFW Operations and is not a part of the Official Record

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

IMAGES ARE BEST AVAILABLE COPY.

As rescanning documents will not correct images, please do not report the images to the Image Problem Mailbox.

*	Consommeters of Corporations Consols Harons des braves	Consumor enco Comporteo Alferio Canada Pottani Olica			un
	California Compania KIA DCO	(31)	(A1)	F,069,602	,027,
		(32)	ı	2992/05/26	£3/3 ₂
		(43)	1	1992/33/20	₽¥

- (51) INTL-CL. CONF-002/04; CONF-002/18; CONF-002/34; CONF-004/642
- (19) (CA) APPLICATION FOR CANADIAN PATENT (12)
- (54) Process for the Preparation of Polyelefine Having a Broad Kolesular Height Distribution
- (72) Winter, Andrews Garmany (Federal Republic of) ; Dolla; Volker - Germany (Federal Republic of) ; Spalock, Walter - Germany (Federal Republic of) ;
- (73) Nonchet Aktiengesellschaft Germany (Fedoral Republic of)
- (DO) (DE) # 41 17 859.0 1991/05/97
- (57) 6 Chains

Socials the specification southined borein as file

DEA 8684 (19 EE)

2069662

HORCER'S ARTTENURBIGLISCHAFT - RUE 91/# 158 Dr. LO/BE

Duscription

25

3Ď

Process for the preparation of polycletins having a broad molecular weight distribution

The is known that motalicrens estalysts in combination with aluminoxames are capable of polymerising platins to give polymerine having a narrow molecular weight distribution (M/M, of 2-3) (J. Folym. Soi., Pol. Chem. Sd. 33 (1985) 2117; ED-A 302 424). Polyolofins of this type with a narrow distribution are suitable, for example, for applications in precision injention andding, injection molding in general and for the production of tibers. For immercus applications, such as, for example, thermoforming, surfaction, blow molding and for the production of polyolofin forms and films, broader or bimodal molecular weight distributions are required.

For polyethylano, it has been proposed to achieve such products by using two or more metallocume catalysts in the polymerication (EP-A 128 845); however, the systems described are achiral catalysts and would give stactic polypropylene on polymematation of propose. However, atactic polypropylene is unsuitable as a structural material.

The preparation of starsoblock polypropylano where N./K. is 13-15 is disclosed in DN-A 3 640 924. These datalyst systems are liberise unsuitable for the formation of polypletine of high tectioity. Furthermore, the metalloceme activities which can be achieved at industrially relevant polymerisation temperatures and the molecular weights of polymer products are too low. In addition, the proposed catalysts give only an exactic polymer at such polymerisation temperatures.

EP-A 310 734 proposes catalyst systems comprising a

- 2 -

misture of a haraccone and a simumouspe for the preparation of polypropylene. Products have bread to bimodel distributions where K./K. is from 3.7 to 19.3

If only the halocone catalyst is used, polypropylane with a broad distribution is obtained at a certain polymerisation temporature, according to SP-A 355 439.

BynGiotectio polygropyleme having a hence distribution is described in ER-A 387 691 (H_{ν}/H_{ν} up to 6.4) if a hainceone is used.

These processes have the common disadvantages of hadring catalyst cross which are too bigh for industrial applications, together with a low polymerisation ectivity, which additionally makes it movesery to carry out thorough, high-cost purification of the property polymer to remove cetalyst residues.

The object was than to find a catalyst system and a process by means of which polyolatine having a broad, bimodal or multimodal distribution can be proposed and which avoid the disadvantages known from the prior art.

The Object is achieved by using a catalyst system comprising at least two stereorigid strouncesnes and on aluminum compound as constalyst.

The invention thus unlates to a process for the proparation of a polyoletin which has a molecular weight distribution K_a/K_a of a 3.0 and which may be monomodal, bisodal or multimodal, by polymerisation or copolymerization of an olefan of the formula R*CH=CHR* in which R* and R* are identical or different and a hydrogen atom or a skyl radical having 1 to 14 carbon atoms, or R* and R*, together with the atoms connecting them, can form a ring, at a temperature of from -60 to 200°C, at a pressure of from 0.5 to 100 bar, in solution, in suspension or in the ges phase, in the pressure of a catalyst

comprising a transition-motal component (metalloceme) and an eluminoseme of the formula II

$$\sum_{k=0}^{n} a_{1} = 0 - \begin{bmatrix} a \\ a_{1} & 0 \end{bmatrix} = a_{1} - a_{2}$$
 (21)

for the linear type and/or of the formula III

for the cyclic type, where, in the formulas II and III, the radicals R may be identical or different and are a C1-C6-mikyl group, a C1-C6-fluoroalkyl group, a C6-C18-. sayl group, a C1-C6-fluoroalyl group or a hydrogen, and n is an integer from 0 to 50, or, instead of the aluminomous, comprises a mixture of an aluminomous of the formula II and/or of the formula III with a compound AiN, which comprises using, as the transition-matal component, at least one sirconocome of the formula I and at least one sirconocome of the formula I and at least one sirconocome of the formula II and at least one

4n which

- and n² are identical or different and are a hydrogen atom, a C₁-C₁₀-alkyl group, a C₁-C₁₀-alkoxy group, a C₆-C₁₀-aryl group, a C₆-C₁₀-aryl group, a C₇-C₄₀-aryl group, a C₇-C₄₀-arylaryl group, a C₇-C₄₀-alkylaryl group, a C₆-C₄₀-arylalkenyl group or a baloger atom.
- and \mathbb{R}^4 are Adontical or different and are a hydrogen atom, a hologen atom, a C_1 - C_1 0-alkyl group, which may be halogenated, a C_2 - C_1 0-aryl group, or a - sin_2 10, - sin_3 10, - sin_3 10 or - pr_2 10 redical, in which \mathbb{R}^{10} is a halogen atom, a C_2 - C_1 0-alkyl group or a C_3 - C_2 0-aryl group.



for \mathbf{g}^k and \mathbf{x}^a , with the provise that \mathbf{x}^p and \mathbf{x}^k are not hydrogen;

=8 R^{11} , =\$1 R^{11} , =\$0., =\$5, =\$0., =\$0., =\$ R^{13} , =\$0., =\$ R^{14} or =\$(0) R^{14} ,

where

 \mathbf{z}^{11} , \mathbf{R}^{12} and \mathbf{R}^{13} are identical or different and are a hydrogen gen atom, a halogen atom, a $\mathbf{c}_1 \cdot \mathbf{c}_{10} \cdot \mathbf{c}_{10} \cdot \mathbf{c}_{10}$

http://patentsl.ic.gc.ca/cache/gif/02069602dis.afp.4.s0.25,r0.gif

6/24/02

which may be halogenated, = C_6 - C_{10} -, preferably C_8 - C_8 -exyl group, a -NR₂¹⁰, -SR¹⁰, -OSIR₃¹⁰, -EXR₃¹⁰ or -PR₂¹⁰ redical in which R^{10} is a halogen atom preferably a observe atom,

2069602

- 7 -

entgrango.

15

20

 H^{ν} is professibly =CR¹¹R¹⁰, ~61R¹²R¹¹, *GeR¹¹R¹¹, *Q-, -8-, =80, *PR¹¹ or =F(D)R¹¹.

R^a and R^d are identical or different and are as defined for R¹⁴.

m and a are identical or different and are zero, 1 or 2, professbly zero or 1, where m plus n is zero, 1 or 2, preferably zero or 1.

20 and R¹⁰ are preferably fluorenyl, indemyl or symbo-10 pentadienyl, it boing possible for these parent straubures also to carry additional substituents as defined for R¹⁰.

Perticularly professed metallocenes are thus those in which, in the formula X, R^1 and R^2 are identical or different and are methyl or oblorine, R^2 and R^4 are hydrogen, R^3 and R^4 are identical or different and are methyl, ethyl or trifluoromethyl, R^7 is a

An Mil section, and a plus a in sect or 1, in

particular the compounds listed in the working exceptes.

of the compounds I mentioned in the working examples, rec-dimothylatily!(2-methyl-1-indenyl); eirconium dichloride, rec-ethylens(2-methyl-1-indenyl); eirconium dichloride, rac-methyl-1-indenyl); eirconium dichloride, rac-methylens(2-methyl-1-indenyl); eirconium dichloride and rec-phenyl(amthyl) silyi(2-methyl-1-indenyl); eirconium dichloride and rec-phenyl(amthyl) silyi(2-methyl-1-indenyl); eirconium dichloride and rec-phenyl(amthyl); eirconium dichloride and rec-phenyl(amthyl-1-indenyl); eirconium dichloride and rec-phenyl(amthyl-1-in

· 55 -

finoroalkyl group, a C_6-C_{10} -aryl group, a C_6-C_{10} -finoroalkyl group, a C_1-C_{10} -alkoxy group, a C_2-C_{10} -alkoxyl group, a C_3-C_{40} -arylalkyl group, a C_8-C_{40} -arylalkyl group, or R11 and R12 or R11 and R13, together with the atoms connecting them, in each case form a ring, and

- H' is silicon, germanium or tin,
- R' and R' are identical or different and are as defined for R''.
- R¹⁴ and R¹⁶ are identical or different and are a monocyclic or polycyclic hydrocambon radical which can form a sandwich structure together with the mirrorium atom, and
- m and n are identical or different and are zero, I or 2, where m plus n is sero, I or 2.

Alkyl is straight-chain or branched alkyl. Halogen (balayemated) refers to fluorine, chlorine, bromine or iodine, preferably fluorine or chlorine.

R1 and R2 are identical or different and are a hydrogen atom, a C₁-C₁₀-, preferably C₁-C₃-alkyl group, a C₁-C₁₀-, preferably C₄-C₁₀-, preferably C₆-C₈: airl group, a C₆-C₁₀-, preferably C₆-C₈: airl group, a C₆-C₁₀-, preferably C₂-C₄-alkenyl group, a C₇-C₄₀-, preferably C₇-C₁₀-airlalkyl group, a C₇-C₄₀-, preferably C₇-C₁₀-airlalkyl group, a C₇-C₄₀-, preferably C₈-C₁₂-airlalkyl group, a C₈-C₄₀-, preferably C₈-C₁₂-airlalkanyl group, or a halogen atom, preferably chloring.

and R^4 are identical or different and are a hydrogen atom, a halogen atom, preferally fluorine, chlorine or browine atom, a C_1-C_{10} , preferably C_1-C_4 -alkyl group, which may be halogenated, a C_6-C_{10} , preferably C_8 - C_8 -eryl group, a $-RR_2^{10}$, $-SR^{10}$, $-OSIR_3^{10}$, $-SIR_2^{10}$ or $-DR_2^{10}$ radical is which R^{10} is a halogen atom professibly a chlorine atom,

2069642

- 6 -

or a $C_1-C_{10}^-$, preferably $C_1-C_2-\pi 1$ kyl group or a $C_4-C_{10}^-$, preferably $C_4-C_4-\pi \gamma 1$ group. S^2 and R^4 are particularly preferably hydrogen.

 R^1 and R^2 are identical or different, preferably identical, and are as defined for R^2 and R^4 , with the proviso that R^3 and R^4 ammed be hydrogen. R^4 and R^4 are preferably (C₁-C₂)-alkyl, which may be halogenested, such as methyl, ethyl, propyl, isopropyl, butyl, isobutyl or triflacromethyl, in particular methyl.

R' is

= $\mathbf{H}(^{11}$, = $\mathbf{A}\mathbf{L}\mathbf{R}^{11}$, = $\mathbf{G}\mathbf{c}$, - $\mathbf{S}\mathbf{c}$, - $\mathbf{G}\mathbf{c}$, - $\mathbf{S}\mathbf{c}$, - \mathbf{S}

preferably C₁-C₄-alkyl group, in particular methyl group, a C₅-C₁₀-fluoroalkyl group, preferably CP₃ group, a C₆-C₁₀-, preferably C₆-C₈-aryl group, a C₆-C₁₀-fluoroaryl group, preferably pontafluorophanyl group, a C₁-C₁₀-, proferably C₁-C₄-alkoxy group, in particular methody group, a C₂-C₁₀-, preferably C₂-C₄-alkenyl group, C₇-C₄₀-, preferably C₇-C₁₀- erylalkyl group, a C₈-C₄₀-, preferably C₈-C₁₂-arylalkanyl group, or a C₇-C₄₀-, preferably C₇-C₁₂-alkylaryl group, or and A¹² or A¹¹ and A¹³, together with the atomic connecting than, in each case form a ring.

M' is milicon, germanium or tim, preferably milicon or

28

Page 1 of 1

2069502

- 8 -

The particularly profound sotallocenes of the formula in are those in which R^1 and R^2 are identical or different and and monthly or obtains,

n+m is sero or 1 and k^{14} and R^{15} are identical or different and are fluoromyl, indemyl or substituted cyclopentactoryl, in particular the compounds of listed in the working examples.

Of particular importance are thus ran-phonyl(swthyl)-= (lyl(indenyl) | sirconium dichloride, diphonylmethylane(9fluoranyl) (dyologontadianyl) sirconius dichioride, isopropylidame(f-fluorenyl)(oyolopentadienyl)girconium dishloride, rec-dimethylatly1(2,3,3-trimethyl-1-oyolupuntadionyl), sirconium dichloride, roc-dimethylsilyl-(indenyl), mirconium dichloride, rwo-dimethylgermylnun-dinathylgormyl-(mitteently strooning dichloride, xao-dimethylsilyl-(indenyl) idimathylsicoonium, ran-phenyl (vinyl) stlyl-(indemyl), sirponium dichleride, rec-H_C-CA,-CH,-Si-(indemy1) seignomies dichloride, rec-dimethy1eily1(2,4dimethylogolopentadienyk), ziroonium dichlozido, racisopropyTidene(indenyl), sirconium dichloride, disethyleilyl(?-methyl-4,5,6,?-totrahydro-1-indenyl);airconium dishlorida, rac-ethylana(indenyl);sirconium · dichloride | Two-methylene(3-t-butyl-1-oyologentadienyl) >zirconium dichloride and rec-dinothylailyi(4,7-dinethyl-1-indemy1) sirconfun dichloride.

The metalloceness having C. symmetry (subgroup of compounds of the formula Is; for example ning C(fluorenyl) -(cyolopentadianyl)dimethylairconium) are employed for the Pumperation of the syndiotectic block in the polyclerin.

For the purposes of the present investion, the term C.

15

20

Page 1 of 1

2069602

symmetry means that the corresponding metallocanes have a mirror plane perpendicular to the plane passing through \mathbb{R}^r , \mathbb{R}^r and \mathbb{R}^s . The bisecting line of the angle \mathbb{C} \mathbb{R}^r - \mathbb{R}^r extends in this mirror plane. This consideration of symmetry is restricted to part of the timescense molecula, i.e. the $-(\mathbb{C} n^r \mathbb{N}^r)_{\mathbb{R}^r} - (\mathbb{C} n^r \mathbb{N}^r)_{\mathbb{R}^r}$ bridge is not taken into abcount. Furthermore, the term \mathbb{C} , symmetry should be understood in formal or identified terms. Thus, for example, shifts in said moisty which may be caused by the bridge and can only be explained via the structure are not considered for the purposes of the present invention.

The chiral metallocomes are employed as recommates for the preparation of highly isotactic polyplefins. However, it is also possible to use the pure N- or 8-form. These pure attractionarie forms allow proparation of an optically series polymer. However, the meso-form of the metallocemes should be removed since the polymerisation-active center (time metal atom) in these compounds is no longer chiral due to mirror symmetry at the central metal and can therefore not produce any highly isotactic polymer. It the meso-form is not removed, etactic polymer is formed alonguide isotactic polymer. For contain applications — soft moldings for example — this say be thoroughly desirable.

25 The principle of resolution of stereoiscens is known.

The metallocones I and Is can be prepared by the principle of the following reaction schome:

x-(cx829), 27-(cx829),-2

- Il -

$$H_{R}^{R} = Q$$

$$M_{R}^{R} = M_{R}^{R}$$
for 1 or 1a

OL

(cf. Journal of Organomet. Chem. (1985) 63-67 and EF-* 320762).

The choice of the motallocenes for the polymerisation of clefins to give polyclefine having a broad or multimodal

2059592

- 10 -

 $\begin{array}{l} {\rm RR}^{G_{*}} \left({\rm CR}^{B} {\rm A}^{9} \right)_{m} {\rm -R}^{7} - \left({\rm CR}^{B} {\rm R}^{9} \right)_{m} {\rm -R}^{d} {\rm K} \\ {\rm Lik}^{G_{*}} \left({\rm CR}^{B} {\rm R}^{9} \right)_{m} {\rm -R}^{7} \cdot \left({\rm CR}^{B} {\rm R}^{9} \right)_{m} {\rm -R}^{d} {\rm Li} \end{array}$

botylli brch

$$\frac{R^{2}L_{\perp}}{R^{2}L_{\perp}} > \frac{(R^{2}R^{2}O)_{R} - R^{2}}{(R^{2}R^{2}C)_{R} - R^{2}}$$

x = Cl, Er, I or 0-tosyl;
$$x_2x^2 = 0$$
 for 7 or $x_2x^3 = 0$

2069602

= 12 stribution can take piece by a

distribution can take piece by means of a test polymorisation for each motallocome.

In this test, the cirlin is polymerized to the polyolefin and the mean molecular weight K, thereof and the unlecular weight distribution K/K, thereof are determined by means of the permeation thromatography. Depending on the desired molecular weight distribution, the metallocular are then combined.

- Taking into account the polymerisation activities, it is then pussible, by means of computer simulation of the combined gol permention curves, to directly produce any desired molecular weight distribution via the type of metallocenes and via the ratio of the amounts of the metallocenes to one another.
- The number of stromogenes to be used according to the invention is graduably 2 or 3, in particular 2. However, it is also possible to use a greater number (such as, for example, 4 or 5) in any desired combination of I and Is.
- By including the polymerication activities and solecular seights at various polymerication temperatures, in the presence of hydrogen as solecular weight regulator or in the presence of commoners, the computer simulation model can be further refined and the applicability of the process according to the invention further improved.
- 25 The countainst used is an aluminomena of the formula II and/or III, where a is an integer from 0 to 50, preferably 10 to 35.

Who radicals R are preferably identical and are methyl, isobutyl, phenyl or benzyl, particularly preferably methyl.

If the radicals R are different, they are preferably mathyl and hydrogen or alternatively mathyl and isobutyl,

- 13 -

hydrogen or isobutyl professibly being present to the extent of 0.01-40% (number of radicals R). The aluminosame can be replaced as cocatalyst in the polymerisation by a nigture comprising aluminosame and $AlR_{\rm c}$, where R is as defined above.

The eluminosano can be prepared in various ways by known promotes. One of the methods is, for example, to react an eluminum hydrocarbon compound and/or a hydridical maining hydrocarbon compound with water (gaseous, solid, liquid or bound - for example as water of crystallization) in an inert solvant (such as, for example, toltabe). To prepare an eluminosano containing different alkyl groups R, two different trialkylaluminum numbounds (AlE, + AlR',), corresponding to the desired composition, are reacted with water (cf. 5. Pasynkiewics, Polyhedron 9 (1990) 429 and EP-A 307 424).

The precise structure of the pluminosames II and III is unknown.

Irrespective of the preparation method, all eluminosane exclutions have in common a varying content of excepted eluminum eterting compound, which is in tree form or on an adduct.

It is possible, before use in the polymerisation resction, to prescrive the metallocenes, in each case esparately or together as a mixture, by means of an aluminousme of the formule (11) and/or (III). This significantly increases the polymorisation activity and improves the particle morphology.

The preactivation of the astalocanes is carried out in solution. The metalicoenes are preferably dissolved, as solution, in a solution of the aluminosane in an inert hydrocarbon. Suitable inert hydrocarbons are alighetic or aromatic hydrocarbons. Toluene or a C₄-C₃₀-hydrocarbon is preferably used.

10

25.

- 14 -

The concentration of the aluminosane in the solution is in the range from about 1% by weight to the saturation limit, preferably from 5 to 30% by weight, in each case based on the total solution. The metalicoenes can be employed in the same commentation, but are preferably employed in an amount of from 10-4-1 make per sole of aluminosans. The prescrivation time is from 5 minutes to 60 hours, preferably from 5 to 60 minutes. The temperature used is from -78°C to 169°C, preferably from 0 to 70°C.

The metallocence way also be propolymerized or applied to a support. Propolymerization is preferably carried out using the (or one of the) elefin(s) suployed in the polymerisation.

associate of suitable supports are silica gols, siminus oxides, solid siminusans or other inorganic support seterials. Another suitable support material is a polyclefic powder in finely divided from.

A further possible embodiment of the process according to
the invention comprises using a salt-like compound of the
formula R_NK_mRR', or of the formula R_FNSK', as covatalyst
in place of or in addition to an eleminosane. In these
formulae, x = 1, 2 or 3, R = alkyl or anyl, identical or
different, and H' = anyl, which may also be fluorinated
or partially fluorinated. In this case, the datalyst
comprises the product of the reaction of the metallocanes
with one of said compounds (of. 52-A 277 004).

In order to remove the catalyst poisons present in the claim, purification by means of an alkylaluminum compound, for example Alms, or Alst, is advantageous. This purification can be carried out either in the polymerisation system itself, or the claim is brought into contact with the Al compound before addition to the polymerisation system and is unbesquently removed again.

10

The polymerisation or copolymerisation is carried out in a known manner in solution, in puspension or in the gas phase, continuously or batchwise, in one or more steps, at a temperature of from -60 to 200°C, preferably from 20 to 80°C. Olefine of the formula R*-CH-CH-R* axe polymerised or copulymerised. In this formula R* and R* are identical or different and are hydrogen stoms or alkyl radicals having 1 to 14 carbon stoms. However, R* and R* may also form a ring with the carbon stoms contacting them, Examples of such elefine are sthylene, propylene, 1-butene, 1-becaus, 4-methyl-1-pentene, 1-cotane, nonhormers and sorbornadiene. In particular, propylene and ethylene are polymerised.

- 15 -

If mnowsery, hydrogen is added as molecular weight regulator. The various hydrogen-reactivities of the metallocanes and the possibility of changing the amount of hydrogen during the polymerisation can result in a turther desired broadening of the molecular weight distribution.

The overall pressure in the polymerization system is from 0.5 to 100 her. The polymerization is preferably carried out in the industrially particularly interesting pressure range of from 5 to 64 her.

The metallocanes are used in a concentration, based on the transition matal, of from 10° to 10° mol, preferably from 10° to 10° mol, of transition motal per dm' of colvent or per dm' of reactor volume. The aluminoxans or the aluminoxans/Alm mixture is used in a concentration of from 10° to 10° wel, preferably from 10° to 10° mol, per dm' of solvent or per dm' of reactor volume. In principle, however, higher concentrations are also possible.

If the polymerication is carried out as a suspension or solution polymerication, an inert solvent which is

10

15

- 16 -

energy for the Singler low-pressure process in user. For example, the polymerization is carried out in an alignatic or cyclocaliphatic hydrocarbon; the examples of these which may be sontioned are butane, pentane, hazane, hoptone, decane, is containe, cycloberane and suthyloyolo-became. It is also possible to use a gasoline or hydrogenated diseal oil fraction. Toluens can also be used. The polymerisation is preferably carried out in the liquid moreoner.

16 Af inert solvents are used, the monomers are metered in in gaseous or liquid form.

The golymerization can take as long as desired, since the catalyst system used according to the invention only axhibits a slight decrease in the polymerization sociuity with time.

The process according to the invention is distinguished by the fact that the metallocanes described give polymers having a broad, bisodal or multimedal molecular weight distribution, high molecular weight, high stereospecificity and good particle morphology in the industrially interesting temperature range between 20 and 90°C with high polymerisation activity.

The polymers according to the invention are particularly suitable for the production of films, in particular transparent files, the motorming applications, polyclaffic fossis, extracion applications and for the production of transparent hollow articles and for blow molding in quadral.

The examples below are intended to illustrate the invertion in greater detail.

the following abbraviations are used:

vs - viscosity number in omi/g

BERNETEL SANGERS

15

20

25

- 17 -

weight average molecular weight in g/mol determined by gel parmmation a molecular weight dispersity apromercal;gobba

II - imotmotic index (MR) + 1/2 MR) Cetermined by syndictactic index **ELECTROSOUPY** (FF + 1/3 MM)

MFI (230/5) = molt flow index, measured in accordance with DIW 53735; molt temperature 230°C and weight 5 kg.

Emample 1 A dry 24 dm³ reactor was flushed with mitrogen and filled with 12 da' of liquid propylene. 39 cm' of a toluene solution of methyleluminomass (sommesponding to 52 maul of Al, mean degree of oligonerisation of the authylaluminomene was n = 19) were then added, and the hatch was stirred at 30°C for 15 minutes.

In parallel, 13.5 mg (0.025 mmol) of rac-phonyl(methyl)dichloride sily1(2-methyl-1-indenyl)stironnium 51.0 mg (0.10 mmol) of rac-phanyl(mathyl)silyl(1indemy1) sirconium dichloride wave dissolved in 15 cm of a toluone solution of methylaluminosana (20 mmol), and the eplution was introduced into the reaster after 15 minutes.

The mixture was polymerized at 30°C for 3 hours. The polymprisation was terminated by addition of 12 1 of CO. 25 gum. 1.85 kg of polypropylene were obtained, corresponding to an activity of the metallocene mixture of 9.6 kg of pp/g of matalloques x h.

 $v_{\rm H} = 341 \text{ cm}^2/\text{c}_{\rm H}$, 411,000 4/201, M,/M = 8.5; II = 96.94. 30

> grandle 2 Example 1 was reposted, but the metallocene mixture components employed were 11.2 mg (0.025 mnol) of ranmthylene(2-methyl-1-indenyl),mirconium shloride 13.9 mg (0.029 mmol) of diphenylmethylene(9-fluoremy1)-(pyolopentadienyl) wlroopinm dichloride;

agreempassing con-

10

13

20

::

- 18 -

polymerization temperature was 60°C and the polymorization time was 1 hour.

2.65 kg of polypropylone were obtained, corresponding to an activity of the motalkocune mixture of 97.6 kg of PF/g of metalkocune \times it.

VH = 295 cm/9) M, = 385,000 g/mol, M/M, = 7.5, The resultant polymer could be separated by fractionation into a fraction of isotactic polymopylane (II > 968) and a fraction of syndictactic polymopylane (SI > 968). The mixing ratio was about 1:1.

Example 3
Example 1 was repeated, but the metallocare mixture components employed were 9.4 mg (0.010 mmn1) of receptory! (methyl) #11y1(2-methyl-1-indemyl) #1 received dishlaride and 8.4 mg (0.013 mmn1) of dimethylmathylene-(9-zimepenyl) (evolopentadionyl) #1 received dishlaride, the polymerisation time was 1 hour.

2.2 kg of a minture of about two parts of inetactic polymographes and one part of syndiotactic polypropylene were obtained, corresponding to an activity of the matallocane mixture of 203.7 kg of FF/q of motallocane x h. vs = 172 cm²/g; K, = 185,500 g/mol, K/K, = 3.0.

Example 4
Example 2 was repeated, but the metallocene mixture
compressed were 4.8 mg (0.01 mm) of racme, 81(2-methyl-1-indenyl) mirroraium dichloxide and 21.2 mg
(0.05 mm) of rec-Me, 81(2,3,5-trimethyloyolopentadienyl) mirroraium dichloride, and the polymerization
temperature was 50°C.

2.57 kg of polypropylene were obtained, nonresponding to an activity of the metallocene mixture of 32.9 kg of PP/g of metallocene \times h.

Я

10

- 19 -

val = 194 cm²/g: M_c = 261,000 g/mul, M_c/M_c = 7.9, II = 98.59.

Example 5

10

Example 1 was repeated, but the metallocate minture components employed were 4.5 mg (0.000 mmol) of recphonyl (methyl) silyl (2-methyl-1-indenyl) sirconium dichloride and 6.6 mg (0.015 mmol) of sac-disathylailyl-(indenyl) sirconium dichloride. The polymerisation time was one hour, and the polymerisation temperature was 50°C.

1.35 kg of polypropylene were obtained, corresponding to an activity of the metallocene mixture of 121.6 kg of PP/g of metallocene \times h.

VM = 156 cm²/g; Km 133,000 g/mol, $P_{\rm c}/M_{\rm h}$ = 8.2, II = 96.0%.

- Example 6
 Example 1 was repeated, but the mutallocens mixture components employed were 2.4 mg (0.005 mmol) of recdiscutyleily1(2-mothyl-1-indenyl); sirconium dighloride
 and 3.5 mg (0.005 mmol) of rec-dimethylermyl(indenyl);sirconium dichloride. The two metallocenes were dissolved
 separately, each in 7.5 cm of a telumna colution of
 methylaluminumnente, and after 15 minutes these solutions
 were metered into the polymerisation system. The mixture
 was polymerized at 70°C for 1 hour.
- 1.57 kg of polypropylene were obtained, corresponding to an activity of the metallocana system of 320.4 kg of PP/g of metallocana \times h.

 VM = 105 cm²/g; K, \times 114,000 g/mol, M,/M, = 4.1, II = 96.35.
- 30 Example 7
 Example 6 was repeated, but the metallocanes used were
 4.8 mg (0.01 mmol) of rec-dimethyleikyl(2-methyl-1indemyl), sinconium dichloride and 1.5 mg (0.004 mmol) of
 rec-dimethylpikyl(indenyl), dimethyleixconium.

- 20 -

2.08 kg f polypropyl no were obtained, corresponding t an activity of the metallocame system of 330.2 kg of PP/g of metallocame \times b. $\forall x = 121 \text{ cm}^2/g$; $x_a = 101,900 \text{ g/mol}$, $x_a/x_a = 4.0$, II = 95.04.

Example 8
Example 5 was represed, but the metallocenes used were
2.7 mg (0.005 mmol) of rec-phenyl(methyl)sklyl(2-mothyl1-indenyl)stroomies dichlorids and 20.5 mg (0.04 mmol)
of mer-phenyl(vinyl)stlyl(indenyl),stroomium dichlorido.

2.17 kg of polypropylene were obtained, corresponding to an activity of the metallocene system of 63.5 kg of PP/g of metallocene x h.

VM = 102 cm/g; K, = 79,400 g/mol, M/M, = 3.5, IX = 96.9%.

- Hemple 9

 Hemple 6 was repeated, but the metallocenes used were

 4.8 mg (0.01 mmol) of reg-dimethylathyl(2-methyl-1indemyl), wirconium dichioride and 9.2 mg (0.02 mmol) of
 rego-k-C-CH₄-CH₅-Si(indemyl), wirconium dichloride.
- 1.82 kg of polypropylene were obtained, corresponding to en activity of the metalloceme mystom of 130 kg of PP/g of metalloceme.
 vg = 145 cm²/g; N_e = 185,300 g/mol, N_e/N_e = 3.5, II = 96.84.
- 25 Stample 10
 Example 5 was repeated, but the metallocanes used were
 3.7 mg (0.005 smol) of rac-pheny2 (methyl)ally1(2-methyll-indenyl); sirconium dichloride and 3.4 mg (0.006 smol)
 of rac-dimethylsily1(2,4-dimethylcyclopentadianyl);sirconium dichloride.
 - 1.31 kg of polypropylene were obtained, corresponding to an activity of the metallocene system of 256.9 kg of FF/G

(0.01 mmol) of rec-dimethylquemyl(indenyl), eliconicul dichlorida were discolved in 15 cm² of methylaluminosana solution (20 mmol of Al, toluenc), and, after 15 minutes, the solution was introduced into the reactor. The measter contents were hearted to 65°C in 3 minutes and polymenteed at this temperature for one hour.

- 22 -

the polymerivation was torminated by addition of 12 1 of 00, gas, success propylers was recoved in gaseous form, and the polymer product was dried at 00° C/100 mber.

- 2.85 kg of polypropylane ware chimined, corresponding to an activity of the metallocane sniwture of 210.5 kg of PP/g of matallocane \times h. VM = 91 cm²/g; \times = 72,000 g/mol, \times /M, = 4.6, II = 96.05.
- Entemple 14

 Promple 1 was repeated, but the estallocomes used wote 1.4 but (0.010 mmpl) of respectyl/methyl/milyl(2-methyl-1-indenyl),wireschipt at cichloride and 27.0 mg (0.016 mmpl) of respective at 27.5 f.f. whereholder indenyl),wireschipt at 1/2-methyl-4.5, f.f. f. whereholder indenyl);wireschipt dishloride, the polymerisation temperature was 30°C, and the polymerisation time was 1.5 house. 3.86 30
 - 1.51 kg of polypropylene were obtained, convesponding to an activity of the matellocone system of 31.1 kg of 37/g of matellocone \times h. VE = 187 cm²/g; M, = 132,500 g/ss1, M./M, = 5.1, EK = 97.61.
- 25

Records 18

Hazample 1 was reposited, but the mptallumenes used work

4.8 mg (0.010 meol) of rac-disethylsilyl(2-methyl-1indemyl), sirconium dichlorido and 7.0 mg (0.017 mmol) of
rec-stbyless(1-indemyl), sirconium dichloride. The
polymeriastion temperature was 80°C and the
polymeriastion duration was 1 bour.

2069602

oone < h. om"/g: M, ~ 189,500 g/mol, W/M, ~ 3.6, lx =

- Example 11

 Wannois 1 was repeated, but the motalicopes used were
 20.9 mg (0.05 mms); of resphenyl(mothyl)silvi(3-mothyl1-indexyl)mirounium dishioride and his mg (0.08 mms);
 of rate-dimethyl-milyl(2,4-dimethyloyolopentadianyl),sisrophium dishioride. The polymerisation time was
 2 howers. 2.32 My of polymergylene wash obtained, commonpometing to an activity of the motalicomes system of
 19.5 My of MD/g of metalicome # h.

 TW = 286 cm²/g: M, = 436,000 g/msl, M/M, = 7.3, XI =
 56.88. 20
- Numerals 12

 Imagels 1 was repeated, but the Metallucenes used wase
 9.0 mg (0.02 macl) of recomprhylethylene(3-mothyl-1indepyl),#imothim distilleries and 9.5 Mg (0.02 macl) of
 recomplimathylene(1-indepyl);sirronium dishlande,
 and the polyperiestion temperature was 50°C, 1.02 Mg of
 polypropylene were obtained, compensating to an extitity
 of the mouniformer system of 26.8 Mg of FF/G of
 metallocene × h.

 VM = 101 cm²/G; M; = 123,000 g/mol, N;/M; = 6.8, 71 =
 91.66. 13 , 25
 - grample 13 A day 24 day resolve was fixed with nitrogen and filled with 24 day (e.t.p.) of hydrogen and 12 day of liquid
 - 18 ml of a columns solution of transhylaluminum (commuss-porting to 26 well of AlMs,) were then added, and the batch was stirred at $40^\circ C$ for 15 minutes.

In parallel, 5.4 mg (0.01 mmol) of rac-phweyl(methyl)-wilyl(2-methyl-1-indonyl):mirconium dichloride and 4.9 mg

Page 1 of 1

1

http://patentsl.ic.go.ca/cache/gif/02069602dis.afp.21.s0.25.r0.gif

DOROTHY WILLIAMS

6/24/02 105-62 64:20 PM

1.30 kg of polypropylana ware obtained, operesponding to An activity of the metallognum system of 127.1 kg of PF/g of metallognum \times h.

- 23 -

Example 16

Skample 1 was repeated, but the metallocenes used were 6.0 mg (0.010 mmol) of rac-diphenylsityl(2-methyl-1-indenyl); virconium dichloride, 6.0 mg (0.013 mmol) of rac-dimethylailyl(1-indenyl); virconium dichloride and 36.0 mg (0.085 mmol) of rac-dimethylailyl(2,3,5-trimathyleyelopentadienyl); virconium dichloride, the polymerization temperature was 40°C and the polymerization duration was 2 hours.

- 1.79 kg of polypropylane were obtained, corresponding to an activity of the metalloceme system of 18.6 kg of PP/g of metalloceme x h.

 VM = 367 cm³/g, M_s = 293,000 g/mol, M_s/M_s = 5.7. II = 98.0%, MFI (230/5) = 24.6 g/10 min.
- 20 Example 17
 A dry 24 dm' reactor was flushed with propplets and filled with 12 dm' of liquid propylets and with 20 ml of a tolushe solution of trimsthylaluminus (convenyonding to 52 smol of Alke,). The batch was stirred at 30°C for 15 sinutes.

In parallel, 3.0 mg (0,005 mmol) of rac-diphenylatlyl(2-methyl-1-indenyl), sirconium (1.chloride, 2.0 mg (0.004 mmol) of rac-dimethylatlyl(2-methyl-1-indenyl), sirconium diohloride and 2.0 mg (0.006 mmol) of rac-dimethylacmyl(1-indenyl), sirconium diohloride were disepted in 20 cm² of methylaluminosame molution (27 mmol of Al, toluene), and, after 15 minutes, the solution was introduced into the resotor. The mixture was polymerized at 65°C for 1.5 hours.

- 24 -

1.99 kg of polymopylane were obtained, a groupoiding to an activity of the metallocave system of 151.4 kg of FP/g of metallocave \times h.

VM = 193 CM^2/G ; M_c = 195,000 G/mol, M_c/K_c \times 5.8, II \times 96.00, NFI (250/5) = 67 G/I0 min.

Example 18

Example 1 was repeated, but the metallocenes used vero 6.0 mg (0.01 mmol) of rea-diphenylsity1(2-methyl-1-indenyl), strated dishloride and 45.0 mg (0.109 mmol) of rec-methyleno(3-t-butyl-1-cyclopentadicsyl), strated dishloride, the polymerization temperature was 60°C and the polymerization duration was 4 hours.

1.63 kg of polypropyleus were obtained, corresponding to an notivity of the metallocene system of 8.0 kg of PP/0 of metallocene \times h. Vg = 358 cm²/g; M_{ν} = 354,000 g/mol, M_{ν}/M_{ν} = 12.5, II = 83.50.

Example 19
Example 1 was repeated, but the matallocenes used were
5.0 mg (0.010 mmol) of rec-diphenylallyl(2-mathyl-1indenyl);sirconium dichloride and 6.0 mg (0.012 mmol) of
rec-dimethylsillyl(4,7-dimethyl-1-indenyl);sirconium
dichloride, the polymerisation temperature was 40°C and
the polymerisation duration was 2 hours.

0.85 kg of polypropylane were obtained, norresponding to an activity of the metallocane system of 35.4 kg of PP/g of metallocane × h.

VM + 324 cm²/g; N_o = 352,500 g/mol, N_o/N_o = 15.5, II = 25.38.

20 Example 20
Example 1 was repeated, but the metallocenes used worse, 0 mg (0.010 smol) of rec-diphonylatlyl(2-methyl-1-indenyl); sirconium dichloride and 7.2 mg (0.016 mmol) of rec-athylege(2-methyl-1-indenyl); sirconium dichloride.

10

- 25 - 2669602

The polymerication temperature was 50°C and the polymerication duration was 2 hours.

1.44 kg of polypropylana were obtained, corresponding to an activity of the metallocone system of 54.6 kg of PP/g of metallocene \times h.

VM = 727 cm²/g; 제, ~ 406,000 g/mol, 제/제, = 8.0, II = 97.18:

Example 21

10

ENGREENING CONTACT OF THE STATE

Example 20 was repeated, but in addition 75 g of athylene were intered in continuously during the polymerisation. The polymerisation temperature was 60°C and the polymerisation time was 1 hour.

1.65 kg of ethylene-propylene copolymer were obtained, normappending to an activity of the metallocene system of 125.0 kg of copolymer/g of motallocene \times h. WE = 291 cm³/g) K, = 387,000 g/mol, M₂/K_n = 7.4; 4.34 ethylene content with ethylene units predominantly incorporated in an isolated wanner (11 U-1998 analysis).

Example 22

Example 21 was repeated, but 300 g of ethylene ware only
added after a polymerisation time of 30 minutes.

1.63 kg of oppolymor were obtained, corresponding to an activity of the metallocome system of 112.9 kg of oppolymor/g of metallocome x h.

yn m 357 cm³g; m, = 449,000 g/mol, M,/K, = 8.8. The polymor product can be separated by fractionation (decape, diethyl ether) into a polypropylene component and an echylene-propylene rebber component.

Ethylene content of the copolymer 18.4%.

20 Example 23
A dry 150 dm² reactor was flushed with nitrogen and filled at 20°C with 80 dm² of a gasoline fraction with the aromation removed and with a boiling range of

- 26 -

100-120°C. The gas space was then flushed free from mitrogen by injecting 2 has of propylane and releasing the pressure, and repeating this sycle four times.

50 l of liquid propylens were added, and 66 cm of a tolumn solution of mothylaluminoxass (commanpowling to 100 mm) of Al, molecular weight 590 g/mol accoming to expanopic determination) were added and the reactor contants were heated to 50°C.

mydrogen was untered in to give a hydrogen content in the gas space of the resctor of 0.11, and this content was then maintained during the entire polymerization time by topping up (monitoring on-line by gas chromatography).

15.3 mg of rec-mothylathylone(2-methyl-1-indenyl):
sirodnium dichloride (0.033 mmol), 6.3 mg of rec-phenyl(methyl)silyl(2-methyl-1-indenyl):sirodnium dichloride
(0.012 mmol) and 7.0 mg of rec-diphenyl=ilyl(2-methyl-1indenyl):sirodnium dichloride (0.012 mmol) were dissolved
in 32 ml of a toluens solution of mathylalumlnousne
(corresponding to 50 mmel of Al) and, after 15 minutes,
the golytion was introduced into the resotor.

The reactor was kept at a polymerisation temperature of \$0°C for 7 bours by cooling, the polymerisation was them terminated by addition of 2 ber of CO, gas, and the polymer formed was separated from the suspension medium in a pressure filter. The product was dried for 24 hours at \$0°C/200 mbar. 16.4 kg of polymer powder, were obtained corresponding to a metallocene sativity of \$1.9 kg of FF/g of metallocene x h.

VM = 208 cm²/g; M = 248.000 g/mol; M/M = 3.4

II = 97.98; MFI (230/5) = 32 g/10 min, M.p., 151°C

The product had the following motherical data:
Modulus of clasticity is tension (in motordance with
DIN 33457-E) 1,430 M/mm²; notohed impact strength (a. in
accordance with DIN 53453) 5 mJ/mm² at 23°C; Isod impact

10

15

20

25

strength (in accordance with ISO 180/1 C) 59 mJ/mm2 et. 23°C and 12 suf/ymb at -10°C; Isod notched impact strangth (powerding to ISO 180/1 A) 3 mJ/mm at 21°C and 2 mJ/mm at -30°C; ball indentation hardness (pressing, conditioned, 358 W) 94 M/mm and ball indentwition bardness (injection molding, 958 M, in accordance with DIM 52486) 75 M/mm".

EXAMPLE 26 Example 23 was repeated but the astallocece mixture comprised 6.3 mg of rec-phrayl(mothyl)sily1(2-mothyl-1inderly1); electric dichlorids (0.012 maps) and 2.5 mg of rag-dimethylsilyl(I-indenyl);sirconium (8.046 mmol). Polymerization was carried out without hydrogen.

The polymerization was complete after 26 hours. 15 18.7 kg of polymer powder were obtained, corresponding to a metalloceme activity of 101.6 kg of YP/g of netalloceme A 8.

VM = 202 cm²/g; K, = 296,000 g/mol; K,K, = 7.9 XI - 96.48; MFI (230/5) = 39 g/10 min; m.p.: 148°C The product had the following mechanical data: Modulus of elasticity in texuion (in addordance with Din 5347-E) 1,280 H/mm2; notohed impact strength (a. in accordance with DIN 53453) 3 mJ/mm pt 22°C; Isod impact atmength (in ecoordance with ISO 180/1 C) 65 mJ/mm2 at 23°C and 11 mJ/mm at 430°C; Isod notohod impact strength (according to ISO 180/1 A) 2 mJ/mm² at 23°C and 2 mJ/mm² at 30°C; bull indentation hardwass 77 M/mm (procesing, ommittioned, 358 N) and 71 H/mm (injection solding, 358 M, in accordance with DIN 33 456). 30

JUL-26-02 04:30 PM DOROTHY WILLIAMS

- 28 -

HOS SE/T 158

THE EMPEDIMENTS OF THE INVESTIGATION IN UNICE AN EXCLUSIVE PROPERTY ON PRIVILEGE IS CLAIMED AND DESCRIPTION AS POLICIES.

has a nolecular weight distribution K/M, of a 3.0 and which may be monomodal, bisodal on subtimodal, by polymeriaction or repolymeriaction of an eletin of the formula R'CR=CRR* in which R' and R' are identical or different and are a hydrogen atom of an R'kyl reduct having I to 14 carbon atoms, or m' and R', together with the atoms connecting tham, one form a ring, at a temperature of from -60 to 200°C, at a pressure of from 0.5 to 100 bar, in solution, in suspension or in the cas phase, in the presence of a catalyst congrising a transition-metal component (metallocaus) and an aluminoments of the formula II

$$\begin{array}{c}
R \\
A1 - O \\
A1 - O
\end{array}$$

$$\begin{array}{c}
R \\
A1 \\
R
\end{array}$$

$$\begin{array}{c}
(XX) \\
AY
\end{array}$$

for the linear type and/or of the formula III

$$\begin{bmatrix} 1 & 1 & 1 \\ 1 & 1 & 1 \end{bmatrix}_{n-2} \tag{ZII}$$

for the cyclic type, whome, in the formulae II and III, the radicals R may be identical or different and are a C_1 - C_6 -siky), group, a C_1 - C_6 -fluoresiky) group, a C_2 - C_6 -fluoresiky group or hydrogen, and n is an integer from D to BO, or, instead of the aluminowane, comprises a mixture of an aluminowane of the formula II and/or of the formula III with a despound also, which comprises using, as the transition-metal component, at least one mirrounceme of the formula I and at least one

http://patents1.ic.gc.ca/cache/gif/02069602clm.afp.1.s0.25.r0.gif

6/24/02

10

15

20

29 -

sirconcome of the formula is on alternatively at least 2 sirconcomes of the formula I

in which

- and κ^2 are identical or different and ero a hydrogen stam, a $c_{\chi^{-1}10}$ -elkyl group, a $c_{\chi^{-1}10}$ -elkyl group, a $c_{\chi^{-1}10}$ -eryl group, a $c_{\chi^{-1}10}$ -eryl group, a $c_{\chi^{-1}10}$ -eryl group, a $c_{\chi^{-1}10}$ -eryl-elkyl group, a $c_{\chi^{-1}10}$ -eryl-elkyl group, a $c_{\chi^{-1}10}$ -eryl-elyl group, a $c_{\chi^{-1}10}$ -eryl-elyl group, a $c_{\chi^{-1}10}$ -eryl-elkyl group, a $c_{\chi^{-1}10}$ -elyl group or a halogen atom.
- R^3 and R^4 are identical or different and are a hydrogen other, a halogen atne, a $C_1-C_{1R}-alky$] ordup, which may be halogenated, a $C_6-C_{1Q}-aryl$ ordup, or a NR_2^{-10} , -96^{10} , $-95iR_3^{-10}$, $-8iR_3^{-10}$ or -98_2^{-10} radionly in which R^{10} is a halogen atom, a $C_1-C_{1Q}-alkyl$ group or a $C_6-C_{1Q}-aryl$ group,
- and R' are identical or different and are as dolined for R' and R', with the provinc that R' and R' are not hydrogen,

79.9

=BR¹³, ΔA), R^{12} , =Qe-, -8 π , -Q-, -8-, PSO, -8Q₃, -9KR¹³, =QCO, ->KR¹³ or -9{Q}, R^{13} .

VENTO

- gli. R^{12} and R^{13} are identical or different and are a hydrogen atom, a hologen atom, a C_1 - C_{10} -sikyl group, a C_2 - C_{10} -rivorantkyl group, a C_3 - C_{10} -rivorantkyl group, a C_4 - C_{10} -sikoxy group, a C_5 - C_{10} -sikoxy group, a C_7 - C_{40} -arylalkyl group, a C_7 - C_{40} -arylalkyl group, a C_7 - C_{40} -arylalkonyl group or a C_7 - C_{40} -arylalkonyl group or a C_7 - C_{40} -arylalkonyl group are a C_7 - C_{40} -arylalkonyl group, a C_7 - C_9 -C
 - M' is ellicon, germanium or tim,
 - R^{α} and R^{α} are identical or different and are as defined for R^{α} ,
 - and R are identical or different and are monogolic or polycyclic hydrocarbon radicals which can form a samurich structure together with the sixonium stam, and
 - m . and n are identical or different and are sero, i or 2, where a plus a is zero, 1 or 2.

- 31 -

2. The process as claimed in claim 1, wherein, in the formula R, R' and R' are identical or different and are mothyl or chiorine, R' and R' are hydrogen, R' and R' are hydrogen, R' and R' are methyl, attyl or trifluoromethyl, R' is a

and m plus m is same of 1.

- 2. The propose as claimed in claim 1 or 2, wherein the compound of the formula I is resuding the posthyl-1-indenyl) sirconium dichloride, resuding the confidence (2-methyl-1-indenyl) sirconium dichloride, resuding the local state of the confidence of the confidenc
 - 4. The process as claimed in one or more of claims I to 3, wherein, in the formule Is, H and R are identical or different and are nethyl or chlorine,

n+m is zero or 1 and n^{24} and R^{16} are identical or different and are fluor-mayl, indemyl or substituted cyclopentadicayl.

5. The process as claimed in one of claims 1 to 4, 25 wherein the compound of the formula Is in

69°d

.. 37 --

rap-ph cyl(m thyi)sllyl(indenyl);slro nium dichioride, diphonylwathylone(9-fluoremyl)(cyclopentadianyl) sirconius dichlorida, lacgropylidene(9floorwyl) (cyclopentadienyl) zirconiwa dichlorida, rno-diacthylailyl(2,3,5-trimethy1-1-oyolopuntadionyl), sirconium dichloride, rec-dimethylsilyl-(indepyl) wiscomium dichloride, rec-dimethylocrayi-(indenyl); wireonium dichlorido, zac-dimethyimilyl-(indemy1),dimethylsizoonium,rsc-phenyl(vinyl)silyl-(indemyl) eliconium dichloride, rec-H,C-CH,-CH,-Si-(indenyl); zirdonium dichloride, rec-dimethyleilyl-[2,4-dimethyloyolopontadienyl]; elroralumdichloridm, was-isogropylidens(indenyl);struentum dichlumide, ran-dimethylailyl(3-mathyl-4,5,6,7-tetrakydro-1rec-ethylonudichloride, indenyl) wiropniam (indenyl), sixuosium dishloride, xuo-methylene(3-tbutyl-1-cyclopentudionyl), eirocnium dichloride or xac-dimethylailyl(4,7-dimethyl-1-indenyl),xiruonium alchloxide.

20 6. The process as claimed in one or more of claims 1 to 5. Wherein propylems is polymerized.

10

HOR 91/5 158

ABSINGLY OF THE DISCLOSURE

Process for the preparation of polypletins having a broad molecular swight distribution

rely elefine having a molecular weight distribution M./Na of a 3 and which may be monomodal, bimodal or multimedal are obtained by polymerization or copolymerization of elefins of the formula RCN-CAR, in which a catalyst system comprising an aluminomous and a transition-metal component (metalloceme) is used, the transition-metal component comprising at least one size movement of the formula I

and at least one ziroomocome of the furnula is or alternatively at least 2 siroomocomes of the formula I.